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The Flexible Carbon Activated Electrodes made from Coconut Shell Waste for Supercapacitor Application

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Abstract. The flexible carbon activated electrode supercapacitor application has successfully prepared from coconut shell based carbon materials that produced by local farmers. The Increasing physical and electrochemical properties of coconut shell carbon has been done by chemical activation using KOH and microwave radiation assisted. A Flexible carbon activated electrodes was produced by by spraying method. Physical and electrochemical properties were tested are the specific surface area and specific capacitance. Extensive surface and optimum capacitance were obtained from a flexible carbon electrodes made from coconut shell based carbon is 194.429 m² / g and 10.55 F / g, respectively.

1. Introduction

Electrochemical double layer capacitors (EDLC) or also known as supercapacitors have become an object of research interest in the area study of energy storage device [1]. The electrodes in this device is the main component that determines the amount of energy and power that can be stored in a supercapacitor cell. Looking for a superior electrode is still in progress today, one way is to select the basic raw materials that can be developed into various forms and modifications pores for carbon electrodes. Carbon materials attract to be studied because the advantage of some properties such as the abundant of materials availability, cheap price, pores are easily controlled and stable in the physical and chemical characteristic [2]. Some carbon materials were produced from waste of agriculture. Coconut based waste materials, such as shell has been studied its compatibility as carbon electrode for supercapacitor application. Several previous studies have reviewed on several sides in the preparation of coconut shell as a supercapacitor electrode. Yang in 2010 showed that carbon activated materials can be made from the coconut shell with high surface area [3]. Geng in 2013 has reviewed the various activation agent to produce a high pore surface area activated carbon from coconut shell [4], Hu in 2014 [5] Jain 2014 [6] has reviewed the use of a polymer electrolyte to the coconut shell based electrodes for device of supercapacitor. Some studies that has been shown the coconut shell can be developed as a potential raw material in the production of supercapacitor electrodes.

The Improvement in energy storage capability and power of the supercapacitor and the excellence competitiveness with other energy storage device can also be done by adding dimension and primacy



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in the shape. Flexible supercapacitor cell will facilitate the use of supercapacitors in many appliances. One way of making the flexible supercapacitor cells can be done using the method of spraying the porous carbon material on a sheet of flexible media. This study will report the manufacture of flexible supercapacitor carbon electrodes from carbon material that produced by coconut farmers through the spraying technique. The production of activated carbon from coconut shell is done with by using chemical activation agent with potassium hydroxide and assisted with physical activation using microwave radiation.

2. Experiment

Coconut shell based carbon (CSC) is obtained from a merchant retail in Panam market, Pekanbaru, Riau Province, Indonesia. Carbon based on the coconut shell is produced by local farmers through conventional carbonization techniques. The process of production of activated carbon was begins with the destruction process of the CSC through manual milling techniques to produce small grain size of 0.5 mm. CSC granules was continued by ball milling process for 20 hours to produce a finer grain size is in the range of 50-100 mm. Furthermore, CSCs were activated by chemical activation using KOH as an activating agent with the composition 1: 3, 1: 4 and 1: 5 in weigh ratio. Differences in the composition of carbon and KOH generate different samples and labelled as A1, A2 and A3 for 1: 3, 1: 4 and 1: 5, respectively, the same procedure has been reported before [7, 8]. The next step of each sample was irradiated using microwaves radiated at a power of 600 watts for 20 minutes. Furthermore, sample A1, A2 and A3 that have been irradiated were labelled A1 + R, + R A2 and A3 + R. The last step in the preparation of activated carbon is washed using distilled water and dried to obtain powder coconut shell activated carbon (ACSC).

A cell supercapacitor was constructed by using some of component such as: a pair of Stainless Steel current collector, a separator using whatman filter paper, a pair of ACSC electrodes, and 1 M of H_2SO_4 as an electrolyte. The process of production an activated carbon electrode includes several steps: first, active carbon powder soaked into 1 M H_2SO_4 solution for 2 x 24 hours. Further, the electrolyte solution and ACSC sprayed on one side of the separator with a distance of 15 cm and the number of sprays 3 times. Spraying carbon electrode was conducted at a pressure of 8 kg / cm^2 . Carbon sheets are then dried at a temperature of 65°C for 5 minutes. The same process is subsequently repeated on the other side of the separator. The last stage, the electrode were dried again and flexible carbon electrode were obtained from coconut shell based ACSC. Furthermore, the electrode is ready for fabrication to be a supercapacitor cells for examination, like Figure 1.

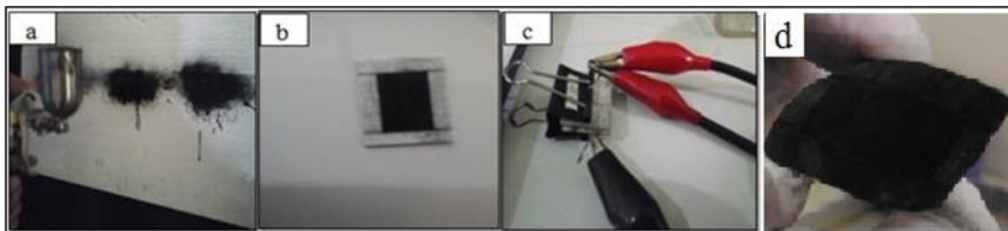


Figure 1. Fabrication of cell supercapacitor: a) process of spraying activated carbon, b) flexible electrode cell, c) flexible supercapacitor cells from coconut shell, d) bent electrode samples showed the flexible characteristic.

Physical properties of ACSC Flexible were performed on the surface morphology and surface area properties. The surface morphology was characterized using a Scanning Electron Microscope (SEM) using a Hitachi S-3400N with a magnification of 10.000 times. ACSC surface area measured by a N_2 gas absorption using the Instrument Nova Station B at a temperature of 77 K. Electrochemical properties supercapacitor cell were carried out by using Cyclic voltammetry (CV) used the handmade

instrument UR RAD-ER 5841 which has been calibrated. The potential window for CV measurement of 0 V to 0.5 V with a scan rate of 1 mV / s and is controlled by software in C ++.

3. Result and Discussion

SEM micrograph images of each of the various samples are shown in Figure 2. SEM micrographs show the difference in colour of dark and light. The differences of brightness level of SEM morphological image may indicate differences in the nature of pores in the particles ACSC. Based on Figure 2, Figure 2b and 2 d which is a sample A3 and A3 + R have the highest brightness level and the lowest. Brightness level morphological differences have relevance to the surface area of the sample.

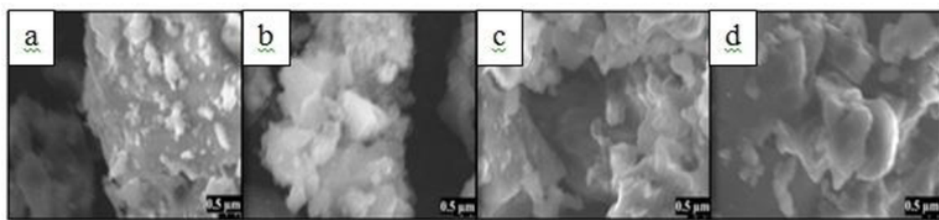


Figure 2. SEM image of coconut shell activated carbon a) A1, b) A3), c) A1+R, d) A3+R

The use of activators KOH on this research has resulted average pores in microporous [9], the pore size of the pore with diameter of ≤ 2 nm. The existence of these pores is indicated by the brightness level morphological SEM lighter. The increasing in the concentration of KOH can increase the number of micropores thus increasing the brightness of SEM micrographs. The power in microwave radiation was produce more light micrograph at A1 + R compared to A1. The process of radiation have resulted in enlarging existing pores and increase the formation of new microporous. On the other hand, the microwave radiation levels uniformly provide better surface regularity on the A1+R and A3 + R. However, the increase of KOH concentration in the samples A3 + R produces SEM micrograph darker. This is due to the formation of a new pore structure on A1 + R better than A3 + R. The increase in the concentration of KOH at A3 + R inhibits the formation of new porous structure by microwave.

CV measurement data obtained in the form of a change of current density vs voltage in cycle curve. The current density is expressed for the two states of charge and discharge condition. Sample data for the CV characteristics measurement are shown in Figure 2. The area formed by the charge and discharge currents have become a key factor in determining the nature of capacitance. The specific capacitation (C_{sp}) of electrode can be calculated using the formula 1, where I_c , I_d , s and m each are: cas current density, discharge current density, the scan rate and the mass of the electrode. The complete results of the charge, discharge for the entire electrode of supercapacitor cells is shown in Table 1. Table 1 shows that the electrode A1 + R has the best value of I_c - I_d . Based on the formula 1 is clearly shown that the specific capacitance is affected by the I_c - I_d and the mass of the electrode. Sample A1 + R shows the highest value of I_c - I_d , this result can be attributed that these sample would yield the highest specific capacitance value. Instead A1 sample showed the lowest value of I_c - I_d . These results suggest that the microwave radiation has changed the nature of the sample pore A1. The formation process flow occurs in a double layer supercapacitors caused by the process of forming a double layer of charge between ions and electrons into the electrode pores. These results clearly show that the samples A1 + R has a pore structure that is easy difused by ion so that the ion-electron pairs more formed.

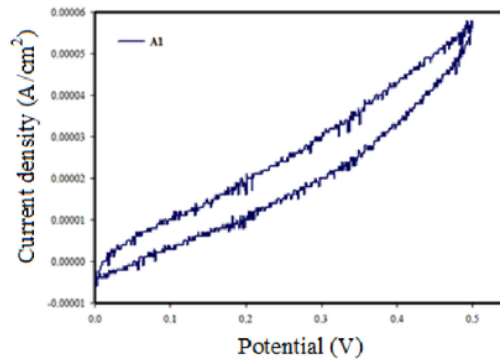


Figure 3. Cv measurement data for A1 sample.

Table 1. Data of CV measurement of coconut based activated carbon supercapacitor electrode.

Samples	Scan rate (V/s)	Mass (g)	Ic (A)	Id (A)	Ic-Id
A1	0.001	0.0165	0.000024	0.000015	0.000009
A2	0.001	0.0190	0.000025	0.000012	0.000013
A3	0.001	0.0175	0.000103	0.000061	0.000042
A1 + R	0.001	0.0165	0.000274	0.000100	0.000174
A2 + R	0.001	0.0180	0.000204	0.000087	0.000117
A3 + R	0.001	0.0170	0.000234	0.000162	0.000072

$$C_{sp} = \frac{(I_c - I_d)}{S \times m} \quad (1)$$

ACSC specific capacitance values correlated to surface area [10]. Specific capacitance value of the electrode and the BET surface area ACSC presented in Table 2. The surface area data of the sample A1 and A3 shows that the increase in carbon and KOH ration, KOH effect on increasing the BET surface area (SBET) [11]. The process of microwave radiation is also shown a role in the increasing in the surface area of the sample.

The increases in the concentration of KOH enhance the Csp and BET surface area on the samples of A1, A2 and A3. Microwave radiation for each various concentrations of KOH activator have been increased the Csp. The A1 sample that have been radiation treatment produce a new pore structure so that the surface area A1 + R larger than A1. Radiation treatment increases SBET [12]. On the other hand, the microwave radiation of 600 Watt to A3 causes a reduction in the BET surface area of the sample A3 + R. This situation is related to the A3 sample has a larger pore size, so the addition of radiation causes the pore size more increases, this situation is causing the decreasing in BET surface area. The Csp for A3 + R sample higher than A3 may influence by surface regularity A3 + R better than A3.

Table 2. The value of specific capacitance and BET surface area of coconut shell based activated carbon electrode.

Sampel	S_{BET} ($\text{m}^2 \cdot \text{g}^{-1}$)	C_{sp} ($\text{F} \cdot \text{g}^{-1}$)
A1	153.815	0.54
A2	-	0.63
A3	200.695	2.40
A1 + R	194.429	10.55
A2 + R	-	6.50
A3 + R	82.755	4.24

Comparison of several flexible supercapacitor electrodes with different precursors are presented in Table 3. This data includes the method of manufacture and the specific capacitance of the flexible electrode. This table shows that the flexible carbon electrodes from carbonized coconut shell by conventionally carbonization process have relatively low capacitive properties than other raw materials. The abundant availability of coconut shell is the advantages of these samples so it certainly can be produced at a relatively low cost. Subsequent studies for physical properties of flexible supercapacitor electrode of coconut shell are expected to increase the electrochemical properties the electrode. Alternative improvement of physical and electrochemical properties still can be more improved such as modification in the carbonization process. Generally, this study has successfully demonstrated that the production of coconut shell based carbon by local farmers as potential materials to be developed as raw materials for a flexible carbon electrode in supercapacitor as an energy storage device.

Table 3. Different flexible supercapacitor electrode from several raw material.

Precursor	Method	C_{sp} (F/g)	Reference
Polyacrylonitrile+ polyvinylpyrrolidone +SiO ₂	Co-electro spinning	242	[13]
Polyethersulfone	Magnetic stirring and ultrasonication	169.4	[14]
TPA + PAN + PVP	Electrospinning	175	[15]
Multi Wall Carbon Nanotubes	Spray method	145	[16]
Activated Carbon Cloth + Polyaniline	Electrodeposition	241	[17]
Carbon Cloth+MnO	Bonding Cu-Wire	425	[18]
Conventional Coconut Shell	Spray method	10.55	Recent study

4. Conclusions

This study has demonstrated the opportunities use of carbon from coconut shell as a flexible activated carbon electrode. The abundant availability, low priced and easily to be activated were good properties of coconut shell based carbon that can be relied upon. Finally, it can be concluded that this study has opened up space in the utilization of carbon material from coconut shell as a potential candidate in the production of activated carbon electrodes for supercapacitor application.

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